

This listing of claims will replace all prior versions, and listings, of claims in the application:

LISTING OF CLAIMS

1. (Previously Presented) A process for the catalytic gas-phase oxidation of propene to acrylic acid, comprising:

passing a reaction gas starting mixture 1 which comprises molecular oxygen and propene in a molar $O_2:C_3H_6$ ratio of ≥ 1 , and at least one inert gas which comprises at least 20% by volume of molecular nitrogen, in a first reaction stage at elevated temperatures, over a first fixed-bed catalyst, whose active material is at least one multimetal oxide comprising molybdenum and/or tungsten and bismuth, tellurium, antimony, tin and/or copper,

thereby obtaining a product gas mixture 1;

wherein said passing of said reaction gas starting mixture 1 proceeds in such a way that a propene conversion in a single pass is ≥ 90 mol%,

an associated selectivity of an acrolein formation and of an acrylic acid byproduct formation together is ≥ 90 mol%,

optionally, a temperature of said product gas mixture 1 leaving said first reaction stage is reduced by indirect and/or direct cooling, and

optionally, molecular oxygen and/or inert gas are/is added to said product gas mixture 1, and

passing said product gas mixture 1, as reaction gas starting mixture 2 which comprises molecular oxygen and acrolein in a molar $O_2:C_3H_4O$ ratio of ≥ 0.5 , and at least one inert gas which comprises at least 20% by volume of molecular nitrogen, in a second reaction stage at

elevated temperatures, over a second fixed-bed catalyst whose active material is at least one molybdenum- and vanadium-containing multimetal oxide,

thereby obtaining a product gas mixture 2,

wherein said passing of said reaction gas starting mixture 2 proceeds in such away that

an acrolein conversion in a single pass is ≥ 90 mol%,

the selectivity of the acrylic acid formation balanced over both reaction stages is ≥ 80 mol%, based on propene converted;

wherein

- a) a loading of said first fixed-bed catalyst with the propene contained in reaction gas starting mixture 1 is ≥ 160 l(S.T.P.) of propene/l of catalyst bed \cdot h,
- b) said first fixed-bed catalyst comprises a catalyst bed arranged in two spatially successive reaction zones A, B,
wherein a temperature of reaction zone A is from 300 to 390°C and a temperature of reaction zone B is from 305 to 420°C and at the same time at least 5°C above the temperature of reaction zone A,
- c) the reaction gas starting mixture 1 flows first through reaction zone A and then through reaction zone B,
- d) the reaction zone A extends to a propene conversion of from 40 to 80 mol%,
- e) a loading of said second fixed-bed catalyst with the acrolein contained in reaction gas starting mixture 2 is ≥ 140 l(S.T.P.) of acrolein/l of catalyst bed \cdot h,

- f) said second fixed-bed catalyst comprises a catalyst bed arranged in two spatially successive reaction zones C,D,
wherein a temperature of reaction zone C is from 230 to 270°C and a temperature of reaction zone D is from 250 to 300°C and at the same time at least 5°C above the temperature of reaction zone C,
- g) the reaction gas starting mixture 2 flows first through reaction zone C and then through reaction zone D, and
- h) the reaction zone C extends to an acrolein conversion of from 55 to 85 mol%.

2. (Original) A process as claimed in claim 1, wherein the reaction zone A extends to a propene conversion of from 50 to 70 mol%.

3. (Original) A process as claimed in claim 1, wherein the reaction zone A extends to a propene conversion of from 65 to 75 mol%.

4. (Previously Presented) A process as claimed in claim 1, wherein the reaction zone C extends to an acrolein conversion of from 65 to 80 mol%.

5. (Previously Presented) A process as claimed in claim 1, wherein the temperature of the reaction zone B is at least 10°C above the temperature of the reaction zone A.

6. (Previously Presented) A process as claimed in claim 1, wherein the temperature of the reaction zone D is at least 20°C above the temperature of the reaction zone C.

7. (Previously Presented) A process as claimed in claim 1, wherein the temperature of the reaction zone B is from 305 to 340°C.

8. (Previously Presented) A process as claimed in claim 1, wherein the temperature of the reaction zone B is from 310 to 330°C.

9. (Previously Presented) A process as claimed in claim 1, wherein the temperature of the reaction zone C is from 245 to 260°C.

10. (Previously Presented) A process as claimed in claim 1, wherein the temperature of the reaction zone D is from 265 to 285°C.

11. (Previously Presented) A process as claimed in claim 1, wherein the propene conversion in a single pass in the first reaction stage is ≥ 94 mol%.

12. (Previously Presented) A process as claimed in claim 1, wherein the selectivity of the acrolein formation and of the acrylic acid byproduct formation together in a single pass in the first reaction stage is ≥ 94 mol%.

13. (Previously Presented) A process as claimed in claim 1, wherein the acrolein conversion in a single pass in the second reaction stage is ≥ 94 mol%.

14. (Previously Presented) A process as claimed in claim 1, wherein the selectivity of the acrylic acid formation balanced over both reaction stages is ≥ 85 mol%, based on propene converted.

15. (Previously Presented) A process as claimed in claim 1, wherein the propene loading of the first fixed-bed catalyst is ≥ 165 l(S.T.P.)/l · h.

16. (Previously Presented) A process as claimed in claim 1, wherein the propene loading of the first fixed-bed catalyst is ≥ 170 l(S.T.P.)/l · h.

17. (Previously Presented) A process as claimed in claim 1, wherein the at least one inert gas contained in the reaction gas starting mixture 1 comprises $\geq 40\%$ by volume of molecular nitrogen.

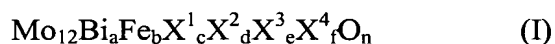
18. (Previously Presented) A process as claimed in claim 1, wherein the at least one inert gas contained in the reaction gas starting mixture 1 comprises $\geq 60\%$ by volume of molecular nitrogen.

19. (Previously Presented) A process as claimed in claim 1, wherein the at least one inert gas contained in the reaction gas starting mixture 1 comprises steam.

20. (Previously Presented) A process as claimed in claim 1, wherein the at least one inert gas contained in the reaction gas starting mixture 1 comprises CO₂ and/or CO.

21. (Previously Presented) A process as claimed in claim 1, wherein the propene content of the reaction gas starting mixture 1 is from 4 to 15% by volume.

22. (Previously Presented) A process as claimed in claim 1, wherein the active material of the first fixed-bed catalyst is at least one multimetal oxide of the formula I



where

X¹ is nickel and/or cobalt,

X² is thallium, an alkali metal and/or an alkaline earth metal,

X³ is zinc, phosphorus, arsenic, boron, antimony, tin, cerium, lead and/or tungsten,

X⁴ is silicon, aluminum, titanium and/or zirconium,

a is from 0.5 to 5,

b is from 0.01 to 5,

c is from 0 to 10,

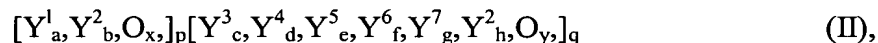
d is from 0 to 2,

e is from 0 to 8,

f is from 0 to 10 and

n is a number which is determined by the valency and frequency of the elements other than oxygen in I.

23. (Previously Presented) A process as claimed in claim 1, wherein the active material of the first fixed-bed catalyst is at least one multimetal oxide of the formula II



where

Y^1 is bismuth, tellurium, antimony, tin and/or copper,

Y^2 is molybdenum and/or tungsten,

Y^3 is an alkali metal, thallium and/or samarium,

Y^4 is an alkaline earth metal, nickel, cobalt, copper, manganese, zinc, tin, cadmium and/or mercury,

Y^5 is iron, chromium, cerium and/or vanadium,

Y^6 is phosphorus, arsenic, boron and/or antimony,

Y^7 is a rare earth metal, titanium, zirconium, niobium, tantalum, rhenium, ruthenium, rhodium, silver, gold, aluminum, gallium, indium, silicon, germanium, lead, thorium and/or uranium,

a' is from 0.01 to 8,

b' is from 0.1 to 30,

c' is from 0 to 4,

d' is from 0 to 20,

e' is from 0 to 20,

f' is from 0 to 6,

g' is from 0 to 15,

h' is from 8 to 16,

x',y' are numbers which are determined by the valency and frequency of the elements other than oxygen in II and

p,q are numbers whose ratio p/q is from 0.1 to 10,

containing three-dimensional regions which are delimited from their local environment as a result of their composition differing from their local environment and have the chemical composition Y_a^1, Y_b^2, O_x , and whose maximum diameters are from 1 nm to 100 μm .

24. (Previously Presented) A process as claimed in claim 1, wherein the first fixed-bed catalyst comprises annular and/or spherical catalysts.

25. (Original) A process as claimed in claim 24, wherein the ring geometry is the following:

external diameter: from 2 to 10 mm,

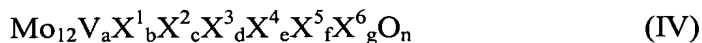
length: from 2 to 10 mm,

wall thickness: from 1 to 3 mm.

26. (Previously Presented) A process as claimed in claim 24, wherein the spherical catalyst is a coated catalyst comprising a spherical support having a diameter of from 1 to 8 mm and a coat of active material applied thereon having a thickness of from 10 to 1000 μm .

27. (Previously Presented) The process as claimed in claim 1, wherein the first and the second reaction stages are each carried out in a two-zone tube-bundle reactor.

28. (Previously Presented) A process as claimed in claim 1, wherein the active material of the second fixed-bed catalyst is at least one multimetal oxide of the formula IV



where

X^1 is W, Nb, Ta, Cr and/or Ce,

X^2 is Cu, Ni, Co, Fe, Mn and/or Zn,

X^3 is Sb and/or Bi,

X^4 is one or more alkali metals,

X^5 is one or more alkaline earth metals,

X^6 is Si, Al, Ti and/or Zr,

a is from 1 to 6,

b is from 0.2 to 4,

c is from 0.5 to 18,

d is from 0 to 40,

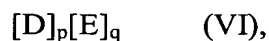
e is from 0 to 2,

f is from 0 to 4,

g is from 0 to 40 and

n is a number which is determined by the valency and frequency of the elements other than oxygen in IV.

29. (Previously Presented) A process as claimed in claim 1, wherein the active material of the second fixed-bed catalyst is at least one multimetal oxide of the formula VI



where

D is $Mo_{12}V_{a''}Z^1_{b''}Z^2_{c''}Z^3_{d''}Z^4_{e''}Z^5_{f''}Z^6_{g''}O_{x''}$,

E is $Z^7_{12}Cu_hH_{i''}O_{y''}$,

Z^1 is W, Nb, Ta, Cr and/or Ce,

Z^2 is Cu, Ni, Co, Fe, Mn and/or Zn,

Z^3 is Sb and/or Bi,

Z^4 is Li, Na, K, Rb, Cs and/or H,

Z^5 is Mg, Co, Sr and/or Ba,

Z^6 is Si, Al, Ti and/or Zr,

Z^7 is Mo, W, V, Nb and/or Ta,

a'' is from 1 to 8,

b" is from 0.2 to 5,

c" is from 0 to 23,

d" is from 0 to 50,

e" is from 0 to 2,

f" is from 0 to 5,

g" is from 0 to 50,

h" is from 4 to 30,

i" is from 0 to 20 and

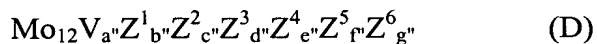
x",y" are numbers which are determined by the valency and frequency of the elements other than oxygen in VI and

p,q are numbers other than zero whose ratio p/q is from 160:1 to 1:1,

which is obtainable by separately preforming a multimetal oxide material (E)



in finely divided form (starting material 1) and then incorporating the preformed solid starting material 1 into an aqueous solution, an aqueous suspension or a finely divided dry blend of sources of the elements Mo, V, Z¹, Z², Z³, Z⁴, Z⁵, Z⁶, which contains the abovementioned elements in the stoichiometry D



(starting material 2), in the desired ratio p:q, drying any resulting aqueous mixture, and calcining the dry precursor material thus obtained, before or after it has been dried, at from 250 to 600°C to give the desired catalyst geometry.

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30. (Previously Presented) A process as claimed in claim 1, wherein the second fixed-bed catalyst comprises annular catalysts.

31. (Previously Presented) A process as claimed in claim 1, wherein the second fixed-bed catalyst comprises spherical catalysts.

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INTERVIEW SUMMARY

Applicants wish to thank Examiner Oh for the helpful and courteous discussion with Applicants' Representative on February 11, 2005. During this discussion it was noted that the claims of U.S. 6, 740,779 fail to disclose or suggest a process for the catalytic gas-phase oxidation of propene to acrylic acid, in which

-in the second stage a **reaction gas starting mixture 2 which comprises molecular oxygen and acrolein and at least one inert gas which comprises at least 20% by volume of molecular nitrogen is used,**

-a loading of said second fixed-bed catalyst with the acrolein contained in reaction gas starting mixture 2 is ≥ 140 l(S.T.P.) of acrolein/l of catalyst bed \cdot h as claimed under e) in Claim 1,

-the reaction zone C extends to an acrolein conversion of from 55 to 85 mol% as claimed in h) of Claim 1.